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Ifelebuegu, A & Johnson, A

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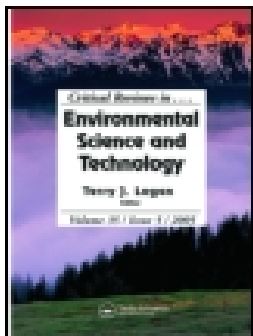
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Non conventional low-cost cellulose and keratin based biopolymeric sorbents for oil/water separation and spill cleanup - A review

Augustine O. Ifelebuegu and Ambulai Johnson

School of Energy, Construction and Environment, Coventry University, Coventry CV1 5FB, UK

A.Ifelebuegu@coventry.ac.uk

Abstract

Oil spills remain an inevitable risk in the exploration, production and transportation of hydrocarbons. Their impacts on the environment are usually severe, hence effective and efficient response strategies are essential to minimise environmental damage. Conventional synthetic oil sorbents are the most widely used sorbents for spill clean-up but are usually expensive and non-biodegradable. Low-cost non-conventional biopolymeric sorbents are now emerging as alternatives to conventional sorbents used in oil spill clean-up owing to their availability and eco-friendliness. In this review, we present low-cost non-conventional cellulose and keratin based biopolymeric oil sorbents highlighting their oil sorption capacities, properties and mechanisms.

Key Words

Biopolymer; Cellulose; Keratin; Oil-spill; Sorbent

1.0 Introduction

Oil and petroleum products play a crucial role in the global economy as they are the key drivers of various industries around the globe. Oil spill pollution is an inevitable risk associated with the exploration, production and transportation of oil. They can cause significant environmental and ecological harm when they occur, and their impact is beyond boundaries and borders. The World has experienced some catastrophic oil spill incidents that have led to significant environmental disasters. Historically, oil spill accidents occurred in different forms and manners owing to the use of different processes of production and transportation. Oil spills are stochastic in nature; however, they occur mainly because of accidents or intentional discharge by man (Etkin, 2004). Oil slicks and intrusion in marine environments can reduce the dissolved oxygen levels in water adversely affecting the wellbeing of the aquatic ecosystem (Ifelebuegu et al. 2017). The feathers of birds and furs of mammals that feed on marine species can also be coated with oil slick, rendering them sick or causing their death (Paul et al., 2013; Beyer et al., 2016).

Oil spill recovery after a spill is a challenging and often a difficult task. The use of inappropriate techniques and measures can cause severe environmental damage. Addressing issues associated with resource management, both materials and humans are vital to developing proactive strategies for containing an oil spill. The availability of financial resources to cover the cost of logistics, besides the availability of trained workforce, remuneration, and support staff, in the attempt to check the devastation is an unavoidable issue. When dealing with oil spills, there is interplay between manpower and materials (Zhong and You 2011). Any material considered as having broad applicability in containing an oil spill besides being cheap must have the requisite

properties of restoring the natural habitat to its original or near-to-original condition in a timely and efficient manner.

Different technologies have been applied in the recovery and remediation of oil spills. They can be classified into physical (mechanical methods and *in situ* burning), chemical treatment, bioremediation and sorption processes (Angelova et al., 2011; Liu et al., 2016). The physical methods involve the use of booms, skimmers and sorbents for containment and recovery. Booms, skimmers, and sorbent materials could be used for oil spill containment, because they serve as barricades controlling the spread of oil slick. These physical methods, which employ mechanical devices, are often associated with high financial costs and the need for logistic support is also high. Booms are unique universal devices used for oil spill containment and recovery. Classified by Ventikos et al. (2004) as a mechanical method, the device can confine the spreading of oil. Dave and Ghaly (2011) classified fence booms, curtain booms, and fire-resistant booms as the three types which are highly reliable in checking the spread of oil. Skimmers are devices that mainly remove oil from the surface of the water while retaining its compositional properties for reuse. Low visibilities, rough windy seas often hamper their use and application in water. *In situ* burning is the cheapest method, but environmental and safety concerns also limit its use.

Chemical treatment involves the use of dispersants or solidifiers for the removal of spilled oil. The process is usually simple and can quickly cover large polluted areas. However, chemical treatment has several limitations. Dispersants can be harmful to the aquatic ecosystem and often ineffective for use with high viscosity and emulsified oil. Also, most spilled dispersible oils have limited dispersion windows limiting application in remote areas. The use of dispersants and

solidifiers is also expensive with high initial and running costs (Liu et al., 2016). Similarly, solidifiers which are usually made from synthetic polymers are expensive and only suitable for removal of small thin oil slicks (Fingas and Fieldhouse, 2011). Bioremediation is another important method for the cleanup of oil spills. It is cheap and usually achieves high removal. It is however ineffective for spills with large coherent mass (Liu et al., 2016), and takes a longer period to achieve proper removal.

Sorbents are materials that are used to remove or recover pollutants either by absorption, adsorption or a combination of both (Karan et al., 2011). Adsorbents function by an external coating of oil onto the surface of the adsorbent; while absorption involves the oil being incorporated internally into the absorbent pores (Figure 1). The effectiveness of absorbents is therefore a function of their porosity. Various organic and inorganic materials have been applied as sorbents for oil spill clean-up. They are also considered one of the most promising solutions for oil spill cleanup due to their simplicity and high efficiency (Abdel-Moghny and Keshawy 2014; Ifelebuegu et al., 2015). For a material to be suitable as an oil spill sorbent, it should possess certain physical characteristics which include oleophilicity, hydrophobicity, high uptake and retention capacity, biodegradability and be inexpensive.

Oil spill sorbents can be classified into three categories namely; natural organic, natural inorganic and synthetic sorbents (Zhu et al., 2011; Abdel-Moghny and Keshawy 2014; Nyankson et al., 2016). Natural inorganic adsorbents have a smaller application in spill clean-up. Their adsorbing capacities are in the range of 4–20 times their weight in oil with little buoyancy (Abdel-Moghny and Keshawy 2014). They include inorganic minerals like aluminas, silicas,

zeolites, clay, alumina-phosphates, and mesoporous adsorbents (Knaebel 2011). Synthetic adsorbents are commercial adsorbents extensively used in oil spill clean-up. The most commonly applied synthetic adsorbents are polystyrene, polypropylene, and polyester foams. They have high hydrophobic and oleophilic properties and can sorb up to 70 times their weight in oil. They can also be used several times after recovery (Abdel-Moghny and keshawy, 2014). These conventional sorbents used in the process of cleaning oil spills are expensive and usually non-biodegradable and hence not environmentally friendly. Conventional sorbents are therefore becoming obsolete because of their high cost, inability to sorb and retain oil adequately, poor biodegradability and recovery. These have led researchers, scientists, and engineers to consider the development of various agricultural and industrial wastes biopolymers as non-conventional materials that are biodegradable, abundant in nature, low-cost and can be used as part of a novel oil spill remediation process. While there have been a few reviews of the applications of different types of sorbents for the removal of oil in spills and wastewater treatment, this paper presents a comprehensive review of the non-conventional low-cost cellulose and keratin based biopolymeric sorbents that have been applied in spill cleanup and oil-water separation both in their natural and modified forms. Their characteristics, mechanisms and oil sorption capacities are highlighted.

2.0 Non-conventional natural sorbents

Many studies in the literature have shown the importance of low-cost adsorbents in dealing with oil pollution. The use of agricultural wastes and by-products from industries in the development of appropriate environmentally sustainable products on a large scale that can replace

conventional adsorbents has been gaining attention since the Deepwater Horizon Disaster (Nyankson et al., 2016). An ideal oil sorbent material in addition to being low-cost and abundant, should have the ability for high oil sorption and retention capacities, low water uptake, good reusability, biodegradability and excellent resistance to both physical and chemical deformations while in use. Biopolymeric materials from agricultural and industrial wastes are gaining attention for these reasons (Ifelebuegu et al., 2015; Liu et al., 2016; Yang et al., 2016,). Low-cost non-conventional biopolymers of cellulosic and keratin origins that have been reported for use in oil water separation and spill clean-up are presented in this review.

2.1 Sorption mechanisms and factors affecting oil sorption.

Sorption of oil to sorbents can happen either by absorption, adsorption or a combination of both. In absorption, there is intermolecular penetration where the oil penetrates the pore spaces of the sorbents. The ability of sorbents to sorb oil by absorption is largely dependent on their porosity and affected by the characteristics of the oil (including viscosity, stickiness and adhesion properties). The transport of oil into the voids of the absorbent material is driven by the capillary forces which can be enhanced by pressure and gravitational forces.

In the case of adsorption; the oil is retained or accumulated on the surfaces without penetrating the sorbent material (Karan et al., 2011). It is usually driven by the attraction between the outer surface of the sorbents and the oil, and is controlled by Van der Waals forces, hydrogen bonding, polarity, steric interactions, hydrophobicity and other physical and chemical interactions (Sabir 2015). The sorption process occurs in three different steps namely; a) oil diffuses to the surface of the sorbent; b) capillary action entraps oil in the sorbent structure and; c) oil agglomerates in

the rough and porous structure of the sorbent (Tansel et al., 2011). It is believed that both absorption and adsorption occur when biopolymeric sorbents are in contact with oil. The oil accumulates instantaneously at the surface of the sorbent through lipophilic interactions or coalescence while absorption occurs through capillary movement into the pores of the sorbent materials (Inagaki et al., 2002; Srinivasan and Viraraghavan, 2008).

The oil sorption capacities of biopolymers are affected by several factors including packing and bulk density of the materials, void volume (porosity), surface morphology, selectivity and oil retention properties of the biopolymers. Studies have also shown that the presences of functional groups like hydroxyl, carboxylic and amino acid groups etc. affect the sorption properties of biopolymers (Wahi et al., 2013; Abdel-Moghny and keshawy, 2014; Ifelebuegu et al., 2015).

3.0 Cellulose based oil sorbents

Cellulose, a major structural material in plant and bacteria cell walls is the most abundant biopolymer in the globe and a primary source of raw materials. Its annual production is estimated at 1.5×10^{12} tons (Abdel-Moghny and keshawy, 2014). It is a biopolymer of anhydroglucose linked by b-glucosidic bonds at the C1 and C4 atoms with three hydroxyl groups at C2, C3 and C6 atoms (Figure 2). It is usually structured into fibrils with a matrix of lignin and hemicellulose (Chen 2014). Cellulosic sorbents are predominantly composed of cellulose, hemicellulose and lignin. While the lignin contents are hydrophobic, the cellulose and hemicellulose contents are hydrophilic and hence often attract a significant amount of water, although it remains sparingly insoluble in water and only partly insoluble in organic solvents. The often low hydrophobicity and buoyancy of natural cellulosic materials is a major limitation for their use in oil water

separations during field applications. Various researchers have made several efforts to increase the hydrophobicity and oleophilicity of cellulose based biopolymers by physical, chemical and thermal modifications. Chemical treatment by acetylation is one of the mostly commonly used modification processes. In this process, the functional groups attached to the cellulose and hemicelluloses are usually replaced by hydrophobic groups through various forms of catalytic and non-catalytic processes. The acetyl group from the acetic anhydride reacts with the hydroxyl group on the cellulose and hemicellulose to form oleophilic acetate groups (O-CO-CH_3). Several studies have shown its effectiveness in improving the hydrophobicity of cellulosic fibres (Teli and Valia, 2013a, 2013b, 2016; Asadpour et al., 2015; Wang et al., 2015). Esterification is also used as a means of increasing the oil sorption and water repellent characteristics of cellulosic biopolymers. The chemical modification causes the replacement of hydroxyl groups by C=O and C-O groups which increases the hydrophobicity and hence oil sorption properties of the fibres (Banerjee et al., 2006; Said et al., 2009). Several other chemical modification methods have also been used including acid treatment (Seyed et al., 2006), surfactant treatment (Ibrahim et al. 2010), and the deposition of metal oxides nanoparticles (Zang et al., 2016). Thermal treatments have also been used to improve the sorption capacities of cellulosic fibers. The commonly used thermal treatment is hydrothermal carbonization in sub or super critical water at high temperatures and pressures (Husseien et al., 2008; Angelova et al., 2011). The treatment often reduces the oxygen containing functional groups and volatile components and increases the carbon content of the cellulose material. Therefore enhancing the hydrophobicity of the hydrothermally prepared sorbents.

In recent years, a new class of highly interconnected porous and low density materials with large specific surface areas and porosity has been fabricated using low cost cellulose biopolymers for applications in oil-water separation. These materials called cellulose aerogels or sponges are a third-generation aerogel succeeding traditional silica and synthetic polymer aerogels (Liu et al., 2016). They are fabricated by replacing liquid within the gels by air with the pore volume or network structure remaining unchanged hence their high porosity (up to 99%), with a heterogeneous pore structure in the inner areas (Figure 3a) and a compact poor structure on the surface (Figure 3b). Cellulose aerogels are easily modified to improve their hydrophobicity and oleophilicity and have hence emerged as a fascinating class of oil absorbers with superhydrophobic and superoleophilic properties. Their oil sorption properties are linked mainly to capillary effects, pore structure and volume, surface wettability, hydrophobic interactions with oil and the effects of van der Waals forces (Jiang et al., 2014; Wan et al., 2015). Cellulose based biopolymers and aerogels that have been reported in the literature for oil spill sorption are presented in Table 1 and discussed in the subsequent subsections.

3.1 Kapok fibre

Kapok tree, widely referred to as “Poor man’s Silk”, is the lightest known fibre tree in the world. Kapok fibre is predominantly composed of cellulose (64%) and lignin (13%) (Reddy and Yang, 2009). It has a large lumen and less dense than water (density of approximately 290 kg/m^3). The kapok fibres have low density, large lumen, and cuticle with the fibre surfaces coated by waxes. These properties give them excellent buoyancy, enabling them to resist water, and hence good

oleophilic and hydrophobic properties, and, do not sink after oil recovery (Abdullah et al., 2010; Karan et al., 2011; Wang et al., 2012).

Several studies on kapok suggest that this sorbent material has a very good oil sorption potential (Hori et al., 2000; Dong et al., 2017). It is considered one of the top biopolymers from agricultural wastes with suitable oil sorption capacity and excellent buoyancy (Rengasamy et al., 2011; Wang et al., 2012). The oil sorption capacity varies with its physical and chemical properties and that of the sorbed oil. Lim and Huang (2007) compared the sorption capacities of kapok fibre and polypropylene. Kapok outperformed polypropylene with adsorption capacities of 36, 43 and 45 g/g for diesel, hydraulic and engine oil respectively at a packing density of 0.02 g/cm³. Abdullah et al. (2010) reported a maximum sorption capacity of 50.8 g/g for engine oil on low density kapok fibre and at low contact angle. Similarly, Rengasamy et al. (2011) reported a high sorption capacity of 52.7 g/g for diesel oil at a contact angle of 44°. Ali et al. (2012) reported sorption capacities of 19.35, 25.71, 60.51 and 49.94 g/g for diesel, crude oil, new engine oil and used engine oil respectively. Additionally, Quek et al. (2015) reported that stirring enhanced the adsorption of palm oil onto kapok fibre with each gram adsorbing more than 200 times its weight in oil.

Wang et al. (2012) investigated the adsorption of oil on raw and modified kapok fibre. For the raw unmodified kapok, a sorption capacity of 38.1 g/g was achieved for diesel and 49.1 g/g for soya bean oil. This was improved to 54.2 and 59.2 g/g when the kapok fibre was modified by silica nanoparticles and hydrolyzed dodecyltrimethoxysilane. In another experiment, Wang et al. (2013b) investigated the sorption of oil by kapok fibre modified by coating with polybutyl-

methacrylate and hydrophobic silica. The reported sorption capacities were 59.5, 64.9, 83.2 and 80.3 g/g for gasoline, diesel, soya bean oil and paraffin oil respectively. The coated fibre maintained its sorption properties for 6 cycles of reuse. The mechanism of removal was a complex combination of chemical interaction and van der Waal forces. Dong et al. (2016) blended equivalent amounts of kapok and PET in an oil water separation experiment. The combination achieved sorption capacities of 63 g/g for vegetable oil and 58.5 g/g for motor oil with kapok maintaining its hollow lumen structure after the modification. In another recent work, Wang et al. (2016a) developed a kapok packed module connected to a vacuum pump for a continuous oil water separation. The assembly was efficient in removal of low viscosity oil from water. Wang et al. (2016b), immobilised Fe_3O_4 on the surface of kapok fibre in fabricating a magnetically superhydrophobic sorbent. The modified kapok fibre showed excellent oil sorption capacity and magnetic responsivity with water contact angle reaching 143° . Sorption capacities of 51, 35.3 and 43 g/g were achieved for the sorption of paraffin oil, gasoline and diesel fuel respectively. The reusability of the sorbent was very high with less than 5% reduction in capacities after 16 cycles of reuse. In a recent work, Dong et al. (2017) prepared a depth filtering systems by an air-laying-bonding method using kapok fibre. At varying packing density, 25.50–32.31 g/g of vegetable oil and 14.42–17.37 g/g of diesel were absorbed by the kapok filters. Centrifugation recovered 80 to 91% of sorbed oils and the fibre retained its capacity without significant drop after 4 cycles of reuse. Cao et al. (2017) also achieved similar sorption capacity of 27.86 g/g for vegetable oil. Zhange et al. (2016) prepared lightweight compressible and multifunctional carbon aerogel using kapok fibre. The prepared aerogel achieved >150 g/g for

gasoline and the authors through a facile process, endowed functionalities like superhydrophobicity and magnetic responsiveness.

The various authors have attributed high sorption capacity to the high porosity, low cellulose content, large bamboo shaped lumens and hydrophobicity of the kapok fibre. Oils are adsorbed on kapok fibre by a combination of hydrophobic interaction and physical forces onto the waxy surfaces. There is also absorption taking place through capillary movement of oils into the lumens (Dong et al., 2015b). The adsorption capacities are dependent on the packing densities of the fibre assemblies. The higher their packing densities, the lower the sorption capacities (Lim and Huang 2007; Dong et al., 2016). Other factors that influenced the sorption capacities are the physical characteristics of the oils particularly the viscosity, contact angle and surface tension. The kapok fibre with its excellent sorption for oil has also demonstrated good potential for recovery and reuse with 4 to 15 cycles of use (Lim and Huang, 2007; Abdullah et al., 2010; Wang et al., 2013; Dong et al., 2017).

3.2 Populous seed fibre

Populous is a species of the cotton poplar tree growing 20 – 30 m in length. The tree is an angiosperm from the *Salicaceae* family, common to the northern hemisphere. It is indigenous to Europe, Southwest and Central Asia, and Northwest Africa. The tree produces strand-like fibres which appear white in colour with micropores. Studies on the chemical composition of the fibres showed that lignocellulose is the principal constituent. The components of lignocelluloses (cellulose, hemicellulose, and lignin) which are organized into microfibrils give structural stability to the plant cell wall. Likon et al. (2013) found that the hair fibers are hollow

hydrophobic microtubes with an external diameter between 3 and 12 μm , average length of 4 ± 1 mm, and average tube wall thickness of 400 ± 100 nm. The solid skeleton of the hollow fibres consists of lignocellulosic material coated by a hydrophobic waxy coating. Several authors have reported the good oil sorption characteristics of the populus fibre. In experiments conducted by Likon et al. (2013), maximum sorption capacities of 182–211 g/g and >100 g/g were reported for heavy motor oil and diesel respectively at a packing density of 0.005 g/cm^3 . The adsorption capacity was found to vary with the packing density of the fibre. Zhang et al. (2014) in another experiment with vegetable oil reported oil sorption capacities of 3.94, 16.76 and 21.57 g/g for unmodified, hydrolysed and acetylated populus fibre respectively. In a more recent work Yang et al. (2017) coated populus fiber with acetylated lignin to prepare oil sorbent. The lignin coated fibre achieved a maximum adsorption capacity of 18.35 g/g for corn oil compared to the raw populus fiber which achieved a sorption capacity of 3.88 g/g. The sorbed oil was recoverable by simple squeezing process and achieved sorption capacity of up to 14.70 g/g after four cycles of reuse.

The super oil sorbent properties of the populus seed fibre can be attributed to its chemical, physical, and microstructural properties (Sabir, 2015). The micro-tubes enable oil entrapment in their empty lumens partly accounting for the very high sorption capacities (Likon et al., 2013). It can also be attributed to the cellulose and lignin content of the hollow fibre. The populus fiber also has very good water repellent properties and excellent buoyancy with water adsorbency of 3.48%, and buoyancy of 90%.

3.3 Cotton fibre

Cotton is a spongy, feathery staple fibre composed of cellulose (91%), water (7.85%), protoplasm pectin (0.55%), waxes, fatty substances (0.4%) and mineral salts (0.2%). Cellulose, which accounts for the largest percentage of cotton, with a thicker lumen in the secondary wall is responsible for the sorption properties of this natural material. Also, contributing to the good oil sorption is the wax on the primary walls of the fibre. This wax layer which contains a mixture of long chain saturated fatty acids, alcohols, resins and sterols gives the cotton fibre its hydrophobicity and hence ability to attract oil by non-polar mechanism (Carmody et al., 2008). Deschamps et al. (2003) investigated the removal of oil (vegetable oil, mineral oil, fuel oil and crude oil) from water using raw and acetylated cotton fibre. They reported sorption capacities in the range of 19 to 23 g/g and 23 to 30 g/g for raw cotton and acetylated cotton respectively. The fibres were recovered by simple squeezing and reused for up to 10 cycles. After the tenth cycle adsorption capacities reduced from 15 to 21 g/g and 11.5 to 12.5 g/g for both raw and treated cotton respectively. Surprisingly, the treated cotton showed lower sorption capacities to the raw cotton fibres. It however, demonstrated higher lipophilic properties and greater stability in aqueous medium over time after the sorption experiments. Hussein et al. (2011) found in their study that low grade cotton fibre and pad demonstrated good potential for oil spill clean-up with sorption capacities ranging from 18.43 to 22.5 g/g. The oil retention capacities for both the loose fibre and pad were found to high also. Similarly, Singh et al. (2013) investigated the oil sorption capacity of low-micronaire raw cotton fiber and reported a maximum sorption capacity of 30.5 g/g. They attributed the high sorption capacities to the morphology of the cotton and the low cellulose content of the immature cotton fibres. In further work, cotton fibres were carded and aligned in a single direction. This resulted in a 65%

improvement in sorption capacity due mainly to the low bulk density of the aligned fibres. Peterson et al. (2016) also investigated the sorption characteristics of cotton and polypropylene mats at room and frozen temperatures. Cotton mat had a maximum sorption capacity of 11.48 and 13.87 g/g compared to propylene which had sorption capacities of 9.82 and 11.29 g/g at room and frozen temperatures respectively. The cotton mats combined both absorption and adsorption process in the sorption of oil while polypropylene was based on the adsorption process.

Carbon fibres with hollow tubular structure were derived from natural cotton by Wang et al. (2013a). They reported sorption capacity of 32 to 77 g/g for various oil types include vegetable and crude oil. The high sorption capacities were attributed to the improved surface area and hydrophobicity. They also demonstrated the reusability of the hollow fibre by recovering and reusing them. After five cycles of reuses, the drop-in sorption capacity was only 9% for the hollow fibre compared to 27% for untreated cotton fibres. Bi et al. (2013) investigated the use twisted carbon fibre (TCF) aerogel made from raw cotton for the sorption of petroleum products and vegetable oils. They reported a very high sorption capacity of 50 to 192 g/g. The TCF aerogel showed high hydrophobicity with all the hydrophilic functional group on the raw cotton fibres eliminated. It also demonstrated excellent reusability after the sorbed oils were recovered by simple distillation and mechanical squeezing. Only about 3.7% reductions in sorption capacities were noticed after 5 cycles of reuse. Hoai et al. (2016) investigated the oil sorption capacity of natural cotton modified by stearic acid using 4-(dimethylamino) pyridine and N, N'-dicyclohexylcarbodiimide as a catalyst and a co-catalyst, respectively. The modification increased the hydrophobicity of cotton with a water contact angle of 141 – 144°. The sorption

capacities for three different oils were 23–26, 21–25, 20–24 g/g for crude oil, cooking oil, motor oil respectively. The modified cotton was used for ten cycles of sorption without a significant drop in sorption capacities.

Zhang et al. (2013) and Liu et al. (2014) also recently reported the fabrication of superhydrophobic and superoleophilic cotton fibres for the oil water separation. Both reports showed excellent oil water separation capabilities of the modified cotton fibre with water contact angles greater than 150° , oil contact angle less than 5° , and sorption capacities of up to 50 g/g. A similar result was achieved by Rana et al. (2016) with a water contact angle of 170° when cotton fibre was modified by a single step alkali treatment. The fibres retained their sorption capacities after several cycles of reuses without significant deterioration. Similarly, Zhou et al. (2013) fabricated a durable superhydrophobic cotton fabric by facile vapour phase deposition of polyaniline and fluorinated alkyl silane. The cotton fabric achieved up to 97.8% oil water separation efficiency and was reusable for up to 30 cycles without significant loss in efficiency. Bi et al. (2013) fabricated carbon fibre aerogel using twisted carbon fibres from raw cotton. The prepared aerogel was applied for the sorption of various oils and organic solvents. The sorption capacities ranged from 50 to 192 g/g with pump oil achieving the highest sorption capacity of 192 g/g. The sorbed oils were recovered by simple distillation and mechanical squeezing and the sorption capacities did not deteriorate after 5 cycles of reuse. Wang et al. (2015b) in another experiment used hydrophobic modified cotton fiber with silica nanoparticles. The sorption capacities for various oils ranged from 36.7 to 62.6 g/g. The modified fibre achieved up to 80 cycles of reuse. Jin et al. (2015b) also achieved excellent oil water separation using cotton fibre

coated with polydimethylsiloxane with a water contact angle of 157° , sorption capacity of 24.82 to 70.8 g/g and over 500 cycles of reuse.

The sorption of oil onto cotton fibers is thought to predominantly occur at the fibre surfaces. However, Carmody et al. (2008) obtained a surface area of $8.2 \text{ m}^2/\text{g}$ and concluded that there is a limited sorption sites on the surface of the raw cotton fibre. Therefore, the high sorption capacities reported in various literature above can be attributed to other mechanisms, particularly interactions between the oils and the wax present in the primary walls of the cotton fiber, uptake through interfibre capillaries between fiber bundles and internal lumen storage. Several other factors other than those related to the oil properties, contributing to the sorption process include molecular arrangement of the fibres and their surface morphologies.

3.4 Cattail pollen fibre

The cattail fibre is derived from the fruits of the cattail plant. It is a lignocellulosic fibre with a high amount of wax of up to 10.64%. The fibre is brittle and short and mainly used as stuffing for heat retention and life preservers (Abdullah et al., 2010; Dong et al., 2015a). Due to its good buoyancy and hydrophobicity, it has been investigated for its oil water separation characteristics (Cui et al., 2014; Dong et al., 2015a).

Khan et al. (2004) investigated the oil sorption and retention capacities of several biomass sorbents which were compared against a commercial polyester fibre. The experiments were conducted using engine oil. Cattail fibre showed the highest sorption capacity of 11.07 g/g compared to 10.08 g/g for the polyester fibre. In another experiment, sorption capacities of 13.4 g/g and 14.6 g/g were reported for the removal of engine oil and vegetable oil respectively by

cattail fiber (Cui et al., 2014). The retention capacities for both oils were above 95% after allowing to drain for 24 hours. They attributed the relatively high sorption capacities to high surface waxiness, hydrophobic and oleophilic properties of the cattail fibre. Similarly, Dong et al. (2015a) investigated the oil sorption capacity of cattail fiber assemblies with 10-20% polypropylene/polyethylene sheath-core composite fibres. A sorption capacity of 11 to 13 g/g and 11 to 14 g/g were achieved for vegetable and motor oil respectively, with 90% retention after 24 hours of dripping. Reusability of up to 10 cycles was achieved without significant loss in sorption capacities. Cao et al. (2016, 2017) investigated the oil sorption characteristics cattail fibre. The result showed similar sorption capacity of 12 g/g for vegetable oil with more than 88% retention after 24 hours. Cattail fibre showed similar hydrophobic characteristic as kapok, with a water contact angle of 132.78°. The high oleophilic and water repellent properties were due to the high hydrophobic waxiness of the fibre surface. The unique bamboo-shaped structure of the cattail fiber also provided large exposed surfaces for oil sorption.

3.5 *Calotropis gigantea* fibre (milkweed)

The name milkweed is derived from the milky sap produced by the species *Asclepiadaceae*. The milky sap consists of latex containing several complex organic compounds. Milkweed also referred to as *calotropis gigantea* (CGF) has an intra-fiber porosity of 0.83 with a low central cellularity making it suitable for oil sorption. The fiber composition is made up of cellulose (61%), hemicellulose (21%), lignin (8-9%), pectin (3%) and waxes (1.8-3%) (Chen et al., 2013). It also contains hydrocarbons, fatty acids, sterols and terpenes. The fibres are hollow with low

density, lightweight and very thin wall structure. Owing to its hollow structure, they possess good buoyancy properties (Karthik and Murugan, 2016).

Karan et al. (2011) reported oil sorption capacity of 40 g/g for the sorption of crude oil by CGF. Rengasamy et al. (2011) conducted a study on polypropylene, kapok, and CGF. They found a combination of these materials resulted in high uptake of oil. For the CGF and kapok, their intra-fiber porosity, high density, and low cellulose were the significant factors that played a vital role in improving their adsorption capacity. CGF had a sorption capacity of 44.3g/g for crude oil. Their porosities are like those of kapok and have good reusability. It is also considered an eco-friendly and biodegradable material.

Zheng et al. (2016a) used CGF as a sorbent for oil water separation. The fibre exhibited excellent hydrophobic and oleophilic properties and achieved sorption capacities of 37.6 and 41.7 g/g for paraffin oil and rapeseed oil respectively. It has similar large lumen like kapok and the comparable oil sorption can be attributed to the structural assembly and composition. The results also showed that the potential for reusability is high with no significant reduction in oil sorption after 10 cycles of reuse. In a further work by Zheng et al. (2016b), the sorption capacity of CGF was greatly enhanced by treatment with perfluorosilane in a simple immersion treatment. The water contact angle improved from 129° to 131° following treatment in perfluorosilane. The treated CGF achieved sorption capacities of 56.8 to 86.5 g/g for the sorption of soybean oil. After 10 cycles of reuse of the treated fiber, the sorption capacity was still as high as 64.9 g/g for soybean oil.

3.6 Rice husk/straw

Rice husk an agricultural waste, is the outermost layer encapsulating the rice grain and usually separated during the milling process. It is yellowish in colour with a concave shape and it makes up about 20% of the paddy grain before separation. The husk is lightweight with a bulk density of 34 to 40 kg/m³ and typically composed of lignin (28.25%, hemicellulose (18.59%) and cellulose (31.13%) (Wang et al., 2015a). Sun et al. (2002) investigated the use of rice straw acetylated with and without a catalyst for the adsorptive removal of machine oil. There was an 11.2% increase in weight after acetylation without catalyst and the sorption capacity ranged from 16.8 to 20 g/g. A 15.4% increase was achieved with 4-Dimethyl-aminopyridine catalyst during the acetylation process with a sorption capacity of 20.9 to 24 g/g. The sorption capacities of the acetylated risk husk were proportional to the degree of acetylation.

Vlaev et al. (2011), pyrolysed rice husk in a fluidised-bed reactor to produce rice husk ash (BRHA) and white rice husk (WRHA). The products were applied for the sorptive removal of crude oil and diesel. BRHA achieved a sorption capacity of 5.02 and 6.22 g/g, while WRHA achieved a capacity of 2.78 and 2.98 g/g for diesel and crude oil respectively at 25°C. BRHA exhibited higher hydrophobicity and buoyancy than WRHA. When rice husk was thermally treated by heating in a muffle furnace under CO₂ flow at 300 to 800°C, Kudaybergenov et al. (2012) achieved a sorption capacity of 15 g/g for crude petroleum. Similar sorption capacity was also achieved by Kenes et al. (2012) for heavy crude oil using thermally treated rice husk.

Uzunov et al. (2012) investigated the impact of slow pyrolysis at a temperature range of 250 to 700°C on the crude oil sorption properties of rice husk. The highest sorption capacity of about 10 g/g was achieved at a pyrolysis temperature of 350°C compared to 6.5 g/g for raw rice husk.

They concluded that oil sorption capacities of the pyrolysed rice husk are influenced more by the porous structure rather than quantity of the superficial functional groups. Similarly, Angelova et al. (2011) investigated the oil sorption capacity of carbonised rice husk pyrolysed at a temperature of 480°C. The sorption capacities of 3.7, 5.5, 6.0, 7.5 and 9.2 g/g were reported for gasoline, diesel, light crude oil, motor oil and heavy crude oil respectively. The penetration of the oils into the sorbent material decreased with increasing bulk density.

Wang et al. (2015a) investigated the oil sorption capacity of acetylated rice husk cellulose sorbent pre-treated chemically with NaOH/H₂O₂ and biologically with *Aspergillus flavus*. The acetylated rice husk achieved a sorption capacity of 19.66 g/g for marine oil in an aqueous medium compared to 8.26 g/g for the non-acetylated rice husk. The acetylated rice husk had improved oleophilicity due to the stronger intermolecular interaction between the oil and incorporated acetyl groups in the rice husk cellulose matrix (Wang et al., 2015a).

3.7 Barley straw

Barley straw is cellulose based agricultural residue possessing a multi-layered structure with a top layer called cuticle. This layer is covered by a thin film of wax making the straw hydrophobic (Wisniewska et al., 2003). Hussein et al. (2008) evaluated the oil sorption characteristics of thermally treated barley straw. The straw was pyrolysed at 200 to 500°C. They achieved a sorption capacity of 5.9 to 7.6 g/g and 8.1 to 9.2 g/g for diesel and heavy oil respectively. The carbonised barley straw showed high hydrophobic properties when used as sorbent pads adsorbing oil at a rate higher than polypropylene commercial pads. The pads made from the carbonised straw were reusable for two cycles while the raw straw pads were reused for

up to four cycles. Hussein (2009) also investigated the sorption capacity of barley straw for the removal of various oil types in sea water. The sorption capacity of the raw straw ranged from 6.5 to 12 g/g, depending on the oil types and viscosities. The sorbed oil was recoverable by simple squeezing, indicating good potentials for reusability.

Ibrahim et al. (2010) investigated the use of surfactant modified barley straw for the removal of emulsified mineral oil in an aqueous medium. The surface of the straw was chemically modified by using a cationic surfactant (hexadecylpyridinium chloride monohydrate). A maximum sorption capacity of 0.519 g/g was achieved at near neutral pH. A similar oil sorption capacity of 0.613 g/g was reported for canola oil by the authors in an earlier work (Ibrahim et al., 2009). Tijani (2016) evaluated the oil sorption capacity of barley straw and other Canadian biomass. Barley straw achieved a maximum sorption capacity of 6.07 g/g for the sorption of motor oil.

3.8 Sugarcane bagasse

Sugarcane Bagasse is a by-product of the processing of cane sugar. It is a lignocellulosic biomass composed of cellulose (42.8%), hemicellulose (25.8%) and lignin (22.1%) (de Albuquerque Wanderley et al., 2013). Several authors have investigated the uses of sugarcane bagasse as potential sorbents for oil removal. Behnood et al. (2013) examined the oil sorption properties of several cellulosic materials including sugarcane bagasse and sugar leave straw. Maximum sorption capacities of 8 and 5.5 g/g were achieved for the sorption of crude oil by the bagasse and leave straw respectively. The sorption capacity of the bagasse increased with the particle size with the highest oil sorption capacity achieved at a particle size of 0.2 mm. The increase was attributed to the higher interfacial area of the smaller particles.

Sun et al. (2004) acetylated sugarcane bagasse using N-bromosuccinimide as a catalyst in a solvent free process to improve the hydrophobicity of the acetylated bagasse. The modified sorbent was used for the removal of machine oil from water. The sorption capacity ranged from 13.5 to 20.2 g/g with the acetylated bagasse remaining unwet with water in the absence of organic solvent and showing 1.4 – 2 times the oil sorption capacities of commercial polypropylene. The oil sorption increased with weight percentage gain following acetylation. Said et al. (2009) compared the oil sorption capacity of raw and chemically modified sugarcane bagasse. The chemical modification was done by grafting raw sugarcane bagasse with a fatty acid to add a hydrophobic envelope to the bagasse architecture. A sorption capacity of 3 g/g was achieved for the grafted bagasse.

3.9 Banana fibre

Banana a lignocellulosic fibre is usually cultivated for its fruits. The fibre is obtained from the pseudo-stem of the plant (*musa Seplentum*) and, it is composed primarily of cellulose (57.64%) hemicellulose (29.05%) and lignin (13.30%) (Teli and Valia, 2013). The fibre remains an underutilised waste from banana cultivation (Mukhopadhyay et al., 2008).

Teli and Valia (2013b) investigated the acetylation of banana fibre to improve the hydrophobicity and oil absorbency. Raw and acetylated fibres were used in the experiments to remove motor oil from water. The sorption capacities ranged from 11.86 to 18.37 g/g for the acetylated fibre compared to 2.10 g/g for the raw fibre. After the first cycle of use of the treated fibre there was a significant drop in sorption capacity due to the possible collapse of the lumen after squeezing. Sathasivam (2010) modified banana fibre with various fatty acids and applied

the modified fibres for adsorption of engine oil/kerosene mixture in water. Sorption capacities of 10.7, 9.58, 8.90 and 7.65 g/g were achieved for fibres modified with oleic acid, stearic acid, castor oil and palm oil respectively. The untreated fibre achieved a maximum sorption capacity of 2.06 g/g. The reusability test showed no significant drop in sorption capacity after the first three reuse cycles but decreased by 15.2% and 23.5% by the 4th and 5th cycles respectively.

In another experiment, Shang et al. (2016) modified banana cellulose fibre by using succinic anhydride in 1-allyl-3-methylimidazolium chloride. The succinylated banana fiber achieved a maximum sorption capacity of 32.12 g/g at a cellulose dose of 0.05 g and temperature of 15⁰C. Recently, Teli and Valia (2016) reported on the grafting of butyl acrylate onto banana fibre for improved sorption of oil. A maximum sorption capacity of 14.15 g/g was achieved. The reusability was similar to those of acetylated banana fibre previously reported by the same authors Teli and Valia (2013).

3.10 Corn stalk/straw fibre

Corn stalk/fibre is an agricultural by-product from corn production. It is composed mainly of cellulose (28 – 30%), hemicellulose (23-28%) and lignin (12-16%). The fibre microstructure has lumens with an average external diameter of 248 µm and several surface plications which confer large surface areas (Li et al., 2012).

Li et al. (2012) investigated the characterisation of acetylated cellulose fibres made from corn straw for the sorption of crude oil, diesel oil and vacuum pump oil in an aqueous medium. The results showed that the acetylated cellulose fibre showed high hydrophobicity and floated on the oil water mixture for several days without sinking. The sorption capacity ranged from 47.28 g/g

to 67.54 g/g for crude oil, 24.38 to 52.65 g/g for diesel oil and 15.56 – 42.53 g/g for vacuum pump oil. The water contact of the acetylated fibre was in the region of 120.65° compared to 51.1° for the raw fibre. Zhang et al., 2011 modified corn stalk using $\text{H}_2\text{O}_2/\text{NaOH}$ for 14 hours. The treatment increased the number of sorption sites on the corn stalk fibre with oil sorption capacity reaching 14.08 g/g from 4.33 g/g for the raw corn stalk. Similarly, Peng et al. (2013) treated corn stalk by enzymatic modification with cellulose in sodium acetate buffer. The modified corn stalk achieved a sorption capacity of 18.47, 16.15 and 27.23 g/g for vegetable oil, diesel oil and crude oil respectively.

Zhang et al. (2016) prepared superhydrophobic/superoleophilic corn straw fiber by covalent deposition of zinc oxide particles on corn straw fibre and subsequent hydrophobic modification by hexadecyltrimethoxysilane. The modified fibre achieved a sorption capacity of 20.4 g/g with a water contact angle of 155° and surface angle of less than 3° . Asadpour et al. (2016) investigated the oil sorption capacity of corn silk modified with oleic acid. The modified sorbent showed a high degree of hydrophobicity and oil sorption capacity with maximum sorption capacities of 10.7 and 11.90 g/g Tapis and Arabian crude oils respectively.

3.11 Pomelo peel

Pomelo is a giant citrus fruit. The peel is an agricultural waste composed of 46.22% cellulose, 18.84% hemicellulose and 10.24% lignin. Pomelo peels make up about 44 to 54% of the pomelo fruit and contains several functional groups including amidogens, carboxyl and hydroxyl groups (Zhu et al., 2017). Chai et al. (2015) investigated the application of raw, acetylated and styrene treated pomelo peel for the removal of diesel and lubricating oil from water. All three pomelo

peels achieved good oil retaining characteristics with less than 30% loss of adsorbed oil. Maximum sorption capacities of 6.39 and 9.04 g/g were achieved for raw pomelo peel; 18.91 and 26.36 g/g for acetylated peel; 16.50 and 19.39 g/g for styrene modified peel, for diesel and lubricating oils respectively. In another experiment, Zou et al. (2016) examined the removal of diesel oil from aqueous medium using magnetic pomelo peel prepared by a solvothermal method. A maximum sorption capacity of 27.98 g/g was achieved for the magnetic peel based on the second order kinetics. The sorbent showed good continuous reusability with 41.8 % reduction in sorption capacity after the tenth cycle of reuse. Zhu et al. (2017) also fabricated carbon aerogel using waste pomelo peels by hydrothermal carbonization, freeze drying and pyrolysis process. The fabricate aerogel showed very good oil sorption capacity for oils and organic solvents with sorption capacities of 5 g/g and 36 g/g for castor oil and sunflower oil respectively. Adsorbed oil was recovered by simple solvent desorption and the sorbent maintained relatively high sorption capacity after five sorption -desorption cycles.

3.12 Coconut fibre

Coconut coir is an agricultural waste obtained from the fibrous mesocarp layer of the coconut fruit. It is composed of cellulose (38%), hemicellulose (28%) and Lignin (32.8%). The high lignin content contributes to its capacity to act as a sorbent for oil and other pollutants (Hasany and Ahmad, 2006; Carott et al. 2007). Nduka et al. (2008) investigated the oil mopping characteristics of raw and activated coir powder. The un-carbonised coir achieved a sorption capacity of 4.4, 3.64, 3.65 and 2.4 g/g compared to the sorption capacities of the carbonised coconut coir of 4.6, 4.26, 3.20 and 2.52 g/g for crude oil, diesel, kerosene and petrol respectively.

Yusof et al. (2015) used coconut coir modified by esterification with fatty acids (oleoyl chloride and octanoate) for sorption of oil. There were increases in the hydrophobicity of the coconut coir following the esterification process. The oleoylated coir fiber had an improved water contact angle of 85.8° compared to 66.9° for the raw coir fibre. Sorption capacities of 9.14, 12.38 and 9.56 g/g were reported for the raw, oleate and octanoate coir respectively.

Teli and Valia (2013a) used acetylated coir fiber for oil sorption. The fibre was acetylated at 100°C in the presence of 1% NBS catalyst. Sorption capacities of 3.5 g/g for the raw coconut coir improved to 15.75 g/g for the acetylated coir with increasing weight percent gain. Similarly, Teli et al. 2016 obtained a maximum sorption capacity of 13.58 g/g for the sorption of crude oil by coir grafted with butyl acrylate. In another work Ifelebuegu and Momoh (2015) investigated the adsorptive removal of vegetable oil and diesel oil using raw and blended coconut coir. They achieved sorption capacities of 7.23 and 6.53 g/g for vegetable oil and diesel oil respectively. The sorption was slightly improved to 8.8 and 7.92 g/g when a 1:1 blend of coconut coir and human hair was used. Raw coconut coir had significantly high water sorption of 6.6 g/g limiting its potential field application.

Yang et al. (2017) fabricated a magnetic oil sorbent using coco peat powder immobilized with Fe_3O_4 nanoparticles with the aid of mussel-inspired polydopamine. The surface was further modified with low-surface energy octydecylamine. The prepared magnetic sorbent showed high hydrophobicity with water contact angle of 135° and oil sorption capacities of 5.91, 5.14, 5.49 and 6.18 g/g for cottonseed oil, paraffin oil, machine oil and silicone oil compared to the raw coco peat powder which achieved sorption capacities of 4.36, 3.53, 3.94 and 4.79 g/g

respectively. The magnetic sorbent also demonstrated excellent recyclability, with only 15.32% reduction in sorption performance after eleven cycles of reuse.

3.13 Oil palm waste

Oil palm (*Elaeis Guneensis*) is the leading oil producing fruit and is found predominantly in West Africa and South East Asia. The disposal of the fruit bunches and other waste after extraction of the palm seeds remains an enormous environmental burden for companies. In recent years, the various waste parts including empty fruit bunch, fronds and leaves have gained attention as an adsorbent for various pollutants due to low cost and sorption potentials. Asadpour et al. (2016) investigated the oil removal capacity of acetylated oil palm fruit bunch fibre. The BET surface areas of the raw and acetylated fibres were 0.40 and 0.35 m²/g respectively. The acetylated fibre achieved a maximum sorption capacity of 6.8 and 7.0 g/g with the buoyancy of 93.7 to 95.3% for the sorption of Tapis and Arabian crude oils respectively. Similarly, Idris et al. (2014) reported an oil sorption capacity of 2.8 g/g for unmodified fruit bunch and in another work; Sidik et al. (2012) used modified palm leaves for crude oil removal achieving a sorption capacity of 1.18 g/g.

3.14 Waste Paper

Paper is a major component of municipal waste. Conversion of waste paper to new paper is the major route for its recycling. However, there is a limit to the recycling with most new paper still made from virgin fibre (Bi et al., 2014). Waste paper has been used for spill cleanup and oil-water separation either in its paper form or as cellulose aerogels.

Nguyen et al. (2013) prepared ultralight weight cellulose aerogel from waste paper. The surface of the aerogel was functionalised with methyltrimethoxysilane to improve its hydrophobicity and oleophilicity. The as prepared aerogel achieved a maximum sorption capacity of 24.4 g/g for the sorption of crude oil. Similarly, Bi et al. (2014) fabricated a lightweight, porous carbon microbelt (MCB) aerogel using waste paper. The MCB aerogel showed high sorption capacities for various oils (56 – 188 g/g) with pump oil achieving the highest sorption capacity of 188g/g. The sorbed oil was recoverable by simple distillation or squeezing. The recovered sorbent was reused for several cycles.

Han et al. (2015) also fabricated a lightweight and porous carbon aerogel from waste newspaper. The sorbent showed excellent hydrophobicity with a water contact angle of 132°. Sorption capacities of 29 to 51 g/g were achieved for the various oils with no deterioration in capacity after five cycles of reuse for gasoline sorption and less than 40% reduction for pump oil. The high sorption capacities can be attributed to the high porous nature of carbon aerogels. Feng et al. (2015) fabricated cellulose aerogel using waste paper and Kymene crosslinker. The hydrophobicity of the aerogel was improved via chemical vapour deposition using methyltrimethoxysilane. The prepared aerogel achieved a maximum sorption capacity of 95.5 g/g for motor oil at a temperature of 50⁰C. Jin et al. (2015b) used waste newspaper to prepare cellulose aerogel and then silanised the surface using trimethylchlorosilane. The prepared aerogel achieved 12 to 22 g/g sorption capacity for various oils and organic solvents. A maximum sorption capacity of 16 g/g was achieved for engine oil.

3.15 Bamboo fibre

Bamboo is a naturally occurring plant found in abundance in most tropical countries. It is made up of cellulose and hemicellulose fibres imbedded in a lignin matrix. Cellulose fibres are aligned along the length of the bamboo and its chemical composition is similar to that of wood but with higher ash and silica contents. Bamboo has been used as a composite material for many years and has also found application in the preparation of adsorbents like activated carbon and cellulose aerogel (Ahmad et al., 2014; Ma et al., 2014; Wan et al., 2015).

Wan et al. (2015) prepared cellulose aerogels from bamboo fibre via a NaOH/PEG chemical treatment and freeze drying processes. The aerogel which was further treated to enhance its hydrophobicity by chemical vapour deposition was applied for the sorption of waste motor oil. A maximum sorption capacity of 20.6 g/g was achieved for the modified aerogel. In another experiment, Yang et al. (2015) used bamboo chopsticks to prepare carbon fibre aerogel for the sorption of oils and organic solvents. The prepared sorbent achieved a water contact angle of 144° with good oil sorption and selectivity. Maximum sorption capacity of up to 129 g/g was achieved for pump oil. Recoverability of sorbed oil was achieved by combustion and mechanical squeezing, and, reusability was good with up to 61.4% original capacity after 6 cycles of reuse for the sorption of gasoline. Jiao et al. (2016) also prepared carbon fibre aerogel using bamboo fibre as the raw material. The prepared aerogel showed excellent hydrophobicity and water repellent characteristics, sorbing various oils and solvents 21 to 51 times the weight of the original sorbent. 38 g/g sorption capacity was achieved for machine oil. There was no significant deterioration after five cycles of reuse.

3.16 Bacteria Cellulose

Certain bacteria genera like *Acetobacter*, *Sarcina ventriculi* and *Agrobacterium* are known to produce cellulose often referred to as bacterial or microbial cellulose (BC). BC exists as a basic structure of fibril that consists of β -1 \rightarrow 4 glucan chain with molecular formula $(C_6H_{10}O_5)_n$. The glucan chains are held together by inter and intra-hydrogen bonding (Ul-Islam et al., 2012). They are often purer than plant cellulose as there do not contain lignin and other impurities found in plant cellulose; and, have a unique nanostructure with interconnected networks of cellulose nanofibres (Chen et al., 2010), ultrafine network architecture, high mechanical strength and water holding capacity (Saibuatong and Phisalaphong, 2010; Castro et al., 2011).

Wu et al. (2013) fabricated carbon nanofibre aerogel from bacteria cellulose pellicles by freeze drying and subsequent pyrolysis step. The prepared aerogel had very low density with excellent sorption capacity, recyclability and selectivity. The sorption capacity ranged from 106 to 312 g/g for various oils and organic solvents. The sorption capacities of 165, 140, 180 and 170 g/g were achieved for soya bean oil, pump oil, gasoline and diesel oil respectively. Sai et al. (2015) also prepared BC aerogel with surface modification using trimethylchlorosilane. The as prepared BC aerogel exhibited excellent superhydrophobicity and oleophilicity with water contact angle of 146.5° . The sorption capacities ranged from 86 to 185 g/g for various oils with excellent recyclability. There was no significant deterioration in the sorption performance after ten cycles of reuse. In another experiment, Stefelova et al. (2017) prepared cellulose nanofiber using BC in the form of chars. The char achieved sorption capacities ranging from 64 to 120 g/g for various oils and organic solvents.

3.17 Other cellulosic sorbents

Sayyed Amin et al. (2015) investigated the oil removal capacity of a natural plant *Azolla folliculoid*. A sorption capacity of 11.29 g/g and 5.22 g/g were achieved for the removal of engine oil and crude oil from seawater respectively at a temperature of 25°C. The higher capacity for engine oil was mainly due to the difference in viscosity. The *Azolla* plant also exhibited good buoyancy remaining afloat after 24 hours.

Koseoglu (2016) investigated the oil removal properties of biotemplated *Luffa* cylindrical from seawater. The *luffa* cylindrical was coated with chitosan and hence acted both as a carrier for chitosan and as a sorbent. A maximum oil sorption capacity of 2.38 g/g was achieved for the coated sorbent. The value decreased with increasing temperature. Abdelwahab (2014) also investigated the oil sorption capacity of raw *luffa*. The raw *Luffa* showed high hydrophobic-oleophilic properties with higher selectivity for oil over water. Maximum sorption capacities of approximately 9 and 12 g/g were achieved for heavy crude oil and diesel oil respectively. It also showed relatively good reusability with more than 50% capacity retained after three cycles of reuse.

Cheu et al (2016) investigated the use of acetylated pineapple leaves for the removal of dissolved crude oil from oil/water solution. The maximum sorption capacities were 0.038 g/g for the raw pineapple leaves and 0.091 g/g for the acetylated leaves.

Nnaji et al. (2016) investigated the oil sorption capacity of *Dacryodes edulis* leaf a plant found abundantly in equatorial and tropical climate. They achieved a sorption capacity of 3.4 and 5 g/g for the raw and acetylated fibres respectively.

Yang et al 2016 investigated the use of *Platanus* (Plane tree) fruit fibre chemically modified by acetic anhydride in the presence of NBS catalyst as potential oil sorbent. The treated fibre exhibited unique tubular structure with good oil sorption and retention capacity. A maximum sorption capacity of 46.41 g/g was achieved for the sorption of machine oil. The reusability was also good with no significant loss in sorption capacity after 8 cycles of use.

Cholan (*Cyperus Longus L.*) has also been investigated as a sorbent for oil/water separation. The plant exhibited high oleophilicity with sorption capacities of 33, 27.5 and 25 g/g at a packing density of 0.04 g/cm³ for the sorption of motor oil (HD40), crude oil and diesel oil respectively. The retention was higher than 94% after draining for one hour (Shadizadeh et al., 2014).

Li et al. (2014), investigated the oil sorption capacity winter melon aerogel prepared by hydrothermal process followed by pyrolysis. The low-density aerogel (0.048 g/cm³) achieved a crude oil sorption capacity of 25 g/g with a water contact angle of 135°. The high sorption capacity for crude oil was attributed to its high hydrophobicity, low density, highly porous structure, and high pore volume following the treatment to reduce its hydrophilic properties and enhance oleophilicity.

Payne et al. (2012) investigated the use of bleached softwood fibre and achieved a sorption capacity of 6.5 g/g. The softwood fibres exhibited high water wettability which reduced the oil sorption capacity. The wettability was attributed to the presence of the carboxylic acid group in the fibre chemical composition.

Yin et al. (2016) prepared cellulose based aerogel from *Eichhornia crassipes* for use in oil sorption. The sorbent was strengthened using a paper-wet crosslinker polyamide

epichlorohydrin resin. The prepared sorbent showed excellent sorption capacity for both oil and organic solvents ranging from 58.06 to 101.14 g/g. Sorption capacities of 63, 77, 81, 88 and 94 g/g were achieved for crude oil, diesel oil, soybean oil, lubricating oil and silicon oil respectively. The sorption capacities remained relatively high after 16 cycles of recovery and reuse. In another similar work by the same authors (Yin et al., 2017), sorption capacities of 77, 92, 80, 106 and 90 g/g were achieved respectively for the aforementioned oils using cellulose aerogel prepared from *Eichhornia crassipes* and polyvinyl alcohol as crosslinker. The water contact angle of the prepared sorbent was 156.7° . The reusability was also similar after 16 cycles of recovery by simple squeezing and subsequent reuse. The high sorption capacities are attributed to the porous nature of the cellulose aerogel.

4.0 Keratinous sorbents

Keratins are structural proteins with contents of intra and intermolecular disulfide crosslinking. They are found in abundance in the integument in various vertebrates. Together with collagen it is the most important biopolymer in animals. Keratinous fibres are classified into soft (epithelial keratins) and hard keratin (trichocyte keratins). They are made up of crystalline intermediate filaments embedded in an amorphous protein matrix (Ghosh and Collie, 2014), exhibiting a hierarchal structure that ranges from nanoscale to centimeter scale. Keratinous materials are usually abundant in nature and exist as feathers, quills, wool, claws, beaks and hair. The various materials exhibit similar primary and secondary protein structure with the polymer chain consisting of a polypeptide backbone formed from the condensation of *L*-amino acid. Keratinous biopolymers have been investigated as potential sorbents for oil-water separation (Radetic 2003,

2008; Ifelebuegu et al., 2015, Ifelebuegu and Chinonyere 2016). Keratinous materials are able to sorb pollutants (including oil based) by a combination of physiosorption and chemisorption. Sorption can take place through the porous network or, the surface of the material or, by chemical interaction with the functional groups like peptides and amino acids present in keratin proteins (Ghosh and Collie, 2014; Ifelebuegu et al., 2015). Hair, wool and chicken feathers have all been investigated as oil sorbents either in their raw form or those that had undergone several treatments by various methods to improve their oleophilicity and hydrophobicity, similar to those discussed in section 3.0 for cellulose. Keratin based biopolymers that have been reported in literature for oil spill sorption and separation are presented in Table 2.

4.1 Human hair

Human hair is an abundant natural keratin fibre waste. Hair predominantly comprises protein and a mixture of other compounds such as cuticle, water, lipids, and trace elements. The average compositions of normal hair are: carbon: 45.2%, oxygen: 27.9%, hydrogen: 6.6%, nitrogen: 15.1%, and sulphur: 5.2%. The end bonds of the amino acid component indicate that the protein components account for its porosity. Additionally, the keratin found in hair is called “hard” keratin. This type of keratin does not dissolve in water and is quite resilient. Keratin is an important, insoluble protein and, it is made up of 18 amino acids. The most abundant of these amino acids is cysteine which gives hair much of its strength. The cuticle and lipid constituents are responsible for its hydrophobic property. Salt bonds are also present in hair; these are weak physical side bonds that can easily be broken by weak alkaline or acid solutions and changes in pH. These bonds can be reformed by normalising the pH level of the hair. Disulfide bonds are

chemical side bonds. These bonds are stronger and fewer than hydrogen and salt bonds and cannot be broken by heat or water. A disulfide bond is joined to the sulphur atoms of two cysteine atoms to create cysteine. Although the disulfide bonds are fewer than the physical bonds, they are vital for the strength and durability of hair. Hair can be classified into three types based on colour: brown, black, and blonde. This unique natural waste has the perfect surface tensile strength and recovery elastic limit that helps in the repulsion of water, thus making it a good candidate for adsorption of oils. A few researches have reported the oil adsorption characteristics of human hair (Jadhav et al., 2011; Ingoles et al., 2014; Ifelebuegu et al., 2015). The factors which result in high oil adsorption and low water uptake of human hair are keratin and the cuticles that are sheltered by a layer of lipid (18-methyleicosanoic). The presence of its medulla, cortex, and dead cells in the cuticle do not allow water uptake, giving it hydrophobic properties (Bhushan and Chen, 2006). Jadhav et al. (2011) investigated the oil removal capacity of black, brown and blond hairs in an aqueous medium. They found that black hair showed the highest adsorption capacity and the removal ability was attributed to the adhesive forces between oil and hair being greater than the water/hair adhesion. Ingoles et al. (2014) also reported the oil sorption of human hair from wastewater. They found that an increase in pH led to a decrease in the sorption capacity of human hair. They showed that the hair sorption capacity of gent's hair was higher (0.014 g/g) than that of lady's hair (0.010 g/g).

Ifelebuegu et al. (2015) investigated the oil adsorption properties of three hair types namely: Type A (Asian origin), Type B (European origin), and Type C (African origin). The results indicated that type C hair had the highest oil adsorption capacity with maximum adsorption capacities of 9.30, 8.10 and 7.92 g/g for crude oil, vegetable oil and diesel respectively. The

sorption was mainly due to the micropores hence Type C hair with rougher cuticles achieved the highest sorption capacity. The reusability was also good with no significant drop in sorption capacity after four cycles of reuse.

4.2 Chicken and avian feathers

The chicken food industry generates over 5 million tons of chicken feathers (CFs) annually as waste by-product which constitutes a significant and costly waste management challenge. CFs consist of about 91% keratin, which are assembled into α -helix, β -sheet and disordered structures (de la Rosa et al., 2008). CFs consists of about 5 to 7 % of the body weight of chickens. The typical chemical composition includes sulphur (2.9%), nitrogen (15.5%), lipids (15.8%) and ash content (1%) Wrześniewska-Tosik et al. (2012). They have been reported as a potential low-cost sorbent for wastewater treatment (Barone and Schmidt, 2006; Aguayo-Villarreal et al., 2011) and oil sorption (Wrześniewska-Tosik et al., 2012; Ifelebuegu and Chinonyere 2016). They have hollow structures of knots and hooks. These hollow structures contribute to the available pore spaces for binding with the oil adsorbates. Their entangled pore structure and rough morphology of sorbents materials are known to enhance oil retention (Ifelebuegu and Chinonyere, 2016).

Ifelebuegu and Chinonyere (2016) investigated the sorption of oil on CFs in oil/water mixture and achieved maximum sorption capacities of 7.7, 6.1 and 4.1 g/g for vegetable oil, crude oil, and diesel fuel respectively. The higher adsorption capacity for vegetable oil and crude oil are attributed to their higher viscosities, causing them to bond with the active sites on the CFs. The adsorption kinetics varied inversely with increasing temperatures and was better described by the pseudo-second-order kinetic model.

Zhou et al. (2014) fabricated keratin sponges from waste pigeon feathers using the inexpensive sodium disulfite as reducing agent. The prepared sponge was used for the sorption of paraffin oil and soybean oil. Maximum sorption capacities of 38.66 and 35.33 g/g were achieved in pure oil medium for the sorption of paraffin and soybean oil respectively. The capacities were however lower at 12.64 and 11.97 g/g in oil/water medium due to poor hydrophobicity of the sponge. Wattie et al. (2016) extracted keratin from chicken feathers using graft copolymerization. The extracted keratin was modified with oleic acid and used in the synthesis of keratinous cryogel. The as prepared cryogel was enhanced by glutaraldehyde crosslinker for use in the sorption of motor oil. A maximum sorption capacity of 10.76 g/g was achieved. The enhanced sorption capacity was attributed to the crosslinking of glutaraldehyde with the hydrophobic groups and the formation of fibrous structure within the protein matrix.

4.3 Wool fibre

Wool texture is also made of keratin having similar chemical composition as human hair. Their physical structure is however, different from human hair as the number of scaled cells is greater and their free edge is indented. This allows texture intertwining, creating swollen that enhances some water sorption. Studies have shown that wool has an affinity for oil and the good performances are associated with the surface roughness, porosity, the presence of grease (lanolin) and waxes on the surface of wool (Radetic 2003).

Rajakovic-Ognjanovic (2008) investigated the factors affecting the removal of machine oil from water using natural wool fibre (NWF) and recycled wool based non-woven material (RWNM). The sorption capacities of NWF and RWNM ranged from 33 - 43 and 15 - 19 g/g respectively.

The sorption capacities were affected by the pH and temperature with the sorption mechanism reported to be by physiosorption. Similarly, Radetic (2003) used RWNM for the sorption of base oil, diesel and crude oil in artificial seawater. Maximum sorption capacities of 14, 13 and 12 g/g were achieved for base oil, diesel oil and crude oil respectively. Treatment of the RWMN with chitosan and low temperature air plasma did not improve oil adsorption as they were found to affect the lanolin and waxes on the surfaces which partly accounted for their oil sorption performance. In a follow-up work by Radetic et al. (2008), they investigated further oil types and achieved sorption capacity in water oil mixtures of 9.62, 11.06, 12.98, 13.16 and 15.80 g/g for diesel oil, crude oil, base oil, vegetable oil and motor oil respectively. The sorbent exhibited excellent buoyancy and reusability with the capacity reduction not exceeding 50% after five cycles of reuse.

4.4 Silk fiber

Silk floss fiber is a natural protein fiber produced by the species *Ceibaspeciosa* formerly known as *Chorisia speciosa* and belonging to the same family as kapok i.e. Bombacaceae. The species is native to the tropical and subtropical forests of South America and capable of withstanding drought and moderate cold conditions. It produces a heteropolysaccharide fibre comprising rhamnose, arabinose, galactose, and uronic acid. The amino-acid bond in these molecules accounts for the strength of the material. It has a thicker cell wall than other fibers, and is approximately hydrophobic as populus seed fibres. According to Cao et al. (2013), silk floss has the following structural composition: fibroin (75%), sericin (30%), and wax, pigments, sugars, and other impurities (5%). The layer of sericin which is close to the fibroin has a poor

solubility in water that results in lower water uptake with a water sorbency of about 3.75%. Excellent buoyancy and low specific gravity (0.62 g/cm^3) help in sweeping oil from water surfaces. In experiments conducted by Annunciado (2005), silk floss demonstrated high hydrophobicity and oleophilicity (oil sorption capacity of 68.7 to 85.5 g/g) with excellent water repellent properties and buoyancy of 100% in both ionised and salt water.

Moriwaki et al. (2009) also investigated the application of silkworm cocoon waste as an oil sorbent material. The silk waste achieved a sorption capacity of 42 – 52 g/g for motor oil and 37 – 60 g/g for vegetable oil. The sorbent showed good buoyancy in both static and dynamic conditions with up to 90% oil recovery after use. The sorption capacity reduced with every cycle of reused but, still achieved up to 15 g/g after 5 cycles of reuse.

5.0 Conclusion and future outlook

In this review, we presented the used of cellulosic and keratinous based biopolymers and their modification products as sorbents for oil water separation and spill cleanup. Sorption capacities, mechanisms and reusability were discussed. Cellulose and keratin based biopolymers are abundant in nature and their use for oil water separation and spill cleanup has been attracting significant interest in recent times. Some of the natural biopolymers have shown better or similar oil sorption capacities to conventional synthetic sorbents used in the industry. The hydrophobicity and oleophilicity of some biopolymers have also been shown to improve significantly with good reusability rates following various physical and chemical treatments. Biopolymeric sorbents are therefore excellent oil water separation alternatives to synthetic

polymer due to their low cost, abundance in nature, eco-friendliness and potential high oil sorption capacities.

There is an increasing research effort in physical and chemical modifications of biopolymers to increase their oil sorption capacities and water repellent properties. It is anticipated that this will continue to grow and more biopolymers would be used as raw materials for the fabrication of low cost superhydrophobic, superoleophilic sponges and aerogels for oil water separation. Effective and efficient post processing, recovery, reuse and disposal of oil-bearing biopolymers remains a challenge for the fabrication and use of the next generation of oil sorbents.

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Table 1- Cellulose based biopolymers their oil sorption capacities

Sorbent	Sorbate	Treatment/Modification	Sorption Capacity (g/g)	References
Kapok	Diesel		36	Lim and Huang (2007)
	Hydraulic oil		43	Lim and Huang (2007)
	Engine oil		45	Lim and Huang (2007)
	Engine oil		50.8	Abdullah et al. (2010)
	Diesel		52.7	Rengasamy et al. (2011)
	Diesel		19.35	Ali et al. (2012)
	Crude oil		25.71	Ali et al. (2012)
	New engine oil		60.51	Ali et al. (2012)
	Used engine oil		49.94	Ali et al. (2012)

	palm oil		>200	Quek et al. (2015)
	Diesel		38.1	Wang et al. (2012)
	Soyabean oil		49.1	Wang et al. (2012)
	Diesel	Silica nanoparticles and hydrolysis with DTMS	54.2	Wang et al. (2012)
	Soyabean oil		59.2	Wang et al. (2012)
	Gasoline	Coated with polybutyl-methacrylate and silica	59.5	Wang et al. (2013)
	Diesel	Coated with polybutyl-methacrylate and silica	64.9	Wang et al. (2013)
	Soyabean oil	Coated with polybutyl-methacrylate and silica	83.2	Wang et al. (2013)
	Paraffin oil	Coated with polybutyl-methacrylate and silica	80.3	Wang et al. (2013)
	Paraffin oil	Kapok immobilises with Fe ₃ O ₄	51	Wang et al. (2016b)
	Gasoline	Kapok immobilises with Fe ₃ O ₄	35.3	Wang et al. (2016b)
	Diesel	Kapok immobilises with Fe ₃ O ₄	43	Wang et al. (2016b)
	Vegetable	Blending with equivalent amount of PET	63	Dong et al. (2016)

	oil			
	Motor oil	Blending with equivalent amount of PET	58.5	Dong et al. (2016)
	Gasoline	Kapok fibre aerogel	>150	Zheng et al. (2016)
	Vegetable oil	air-laying-bonding	25.5 to 32.31	Dong et al. (2017)
	Diesel	air-laying-bonding	14.42 - 17.37	Dong et al. (2017)
Populus seed fibers (Populus nigra italic)	Heavy motor oil		182–211	Likon et al. (2013)
	Diesel oil		>100	Likon et al. (2013)
	Corn oil		3.94	Zhang et al. (2014)
	Corn oil	Hydrolysis		Zhang et al. (2014)
	Corn oil	Acetylation	21.57	Zhang et al. (2014)
	Corn oil	Coating with acetylated lignin	18.35	Yang et al (2017)
Cotton Fibre	Vegetable oil	Raw (Acetylation)	30 (20)	Deschamp et al. (2003)
	Mineral oil	Raw (Acetylation)	24 (20)	Deschamp et al. (2003)
	Fuel oil	Raw (Acetylation)	23 (19)	Deschamp et al. (2003)

	Crude oil	Raw (Acetylation)	26 (20)	Deschamp et al. (2003)
	Used lub oil		18.43 - 22.5	Hussein et al (2011)
	Motor oil		30.5	Singh et al. (2013)
	Bakken crude oil		11.48 - 13.87	Peterson et al. (2016)
	Vegetable oil	Raw (carbonisation)	34.1 (44.3 - 57.1)	Wang et al (2013)
	Crude oil	Raw (carbonisation)	56.0 (75.4 - 78.9)	Wang et al (2013)
	pump oil	Raw (carbonisation)	30.3 (39.3 - 44.6)	Wang et al (2013)
	various oils	Pyrolysis to produce aerogel	50-192	Bi et al. (2013)
	Crude oil	Stearic acid modification	23-26	Hoai et al. (2016)
	Cooking oil	Stearic acid modification	21-25	Hoai et al. (2016)
	Motor oil	Stearic acid modification	20-24	Hoai et al. (2016)
	Crude oil	Raw (Immobilisation of Silica nanoparticle)	34.7 (52.6)	Wang et al. (2015b)
	Linseed oil	Raw (Immobilisation of Silica nanoparticle)	38 (62.6)	Wang et al. (2015b)
	Paraffin oil	Raw (Immobilisation of Silica nanoparticle)	36 (61.8)	Wang et al. (2015b)

Cattail Pollen Fibre	Engine oil		11.07	Khan et al. (2004)
	Engine oil		13.4	Cui et al. (2004)
	Vegetable oil		14.6	Cui et al. (2004)
	Vegetable oil	10-20% polypropylene/polyethylene	11.0 - 13.0	Dong et al. (2015a)
	Motor oil	10-20% polypropylene/polyethylene	11.0 - 14.0	Dong et al. (2015a)
	Vegetable oil		12	Cao et al. (2016)
Calotropis gigantea (Milkweed)	Crude oil		40	Karan et al. (2011)
	Crude oil		44.3	Rengasamy et al (2011)
	Parafin oil		37.6	Zheng et al. (2016a)
	vegetable oil		41.7	Zheng et al. (2016a)
	vegetable oil	Perfluorosilane immersion treatment	56.8 - 86.5	Zheng et al. (2016b)
Rice Husk/Straw	Machine oil	Acetylation without catalyst	16.8 - 20	Sun et al. (2002)
	Machine oil	Acetylation with 4- dimethyl-aminopyridine	20.9 - 24	Sun et al. (2002)

	Diesel oil	Pyrolysis	2.78 - 5.02	Vlaev et al. (2011)
	Crude oil	Pyrolysis	2.98 - 6.22	Vlaev et al. (2011)
	Crude oil	Thermal treatment under CO ₂	15	Kudaybergenov et al. (2012)
	Marine oil	acetylation and further treatment with NaOH/H ₂ O ₂ with <i>Aspergillus flavis</i>	19.66	Wang et al. (2015)
	Marine oil		8.26	Wang et al. (2015)
	Crude oil	Pyrolysis	10	Uzunov et el. (2012)
	Gasoline	Pyrolysis	3.7	Angelova et al. (2011)
	Diesel oil	Pyrolysis	5.5	Angelova et al. (2011)
	Light crude oil	Pyrolysis	6	Angelova et al. (2011)
	Motor oil	Pyrolysis	7.5	Angelova et al. (2011)
	Heavy crude oil	Pyrolysis	9.2	Angelova et al. (2011)
Barley Straw	Diesel oil	Pyrolysed	5.9 - 7.6	Husseien et al. (2008)
	Heavy	Pyrolysed	8.1 - 9.2	Husseien et al.

	Crude oil			(2008)
	Crude oil		6.5 - 12	Hussein et al. (2009)
	Marine oil	Surfactant modification	0.519	Ibrahim et al. (2010)
	Canola oil		0.613	Ibrahim et al. (2009)
	Motor oil		6.07	Tijani 2016
Sugarcane Bagasse	Crude oil		8	Behnood et al. (2013)
	Machine oil	N-Bromosuccinimide	13.5 - 20.2	Sun et al. (2004)
	Crude oil		3	Said et al. (2008)
Banana Fibre	Motor oil	Acetylation	11.86 - 18.37	Teli and Valia 2013)
	Motor Oil		2.1	Teli and Valia 2013)
	Engine oil/kerosene		2.06	Sathasivam 2010
	Engine oil/kerosene	Oleic acid modification	10.7	Sathasivam 2010
	Engine oil/kerosene	Stearic acid modification	9.58	Sathasivam 2010
	Engine	Castor oil modification	8.9	Sathasivam 2010

	oil/kerosene			
	Engine oil/kerosene	Palm oil modification	7.65	Sathasivam 2010
	Crude oil	Succinylation	32.12	Shang et al. (2016)
	Crude oil	Butyl acrylate grafting	14.145	Teli and Valia (2016)
Corn Stalk/Straw Fibre	Crude oil	Acetylation	47.28 - 67.54	Li at al. (2012)
	Diesel oil	Acetylation	24.38 - 52.65	Li at al. (2012)
	Vacuum pump oil	Acetylation	15.56 - 42.53	Li at al. (2012)
	Crude oil	H ₂ O ₂ and NaOH	14.08	Zhang et al. (2011)
	Vegetable oil	Enzymatic modification in sodium acetate	18.47	Peng et al. (2013)
	Diesel oil	Enzymatic modification in sodium acetate	16.15	Peng et al. (2013)
	Crude oil	Enzymatic modification in sodium acetate	27.23	Peng et al. (2013)
	Crude oil	Zinc oxide deposition and treatment with Hexadecyltrimethoxysilane	20.4	Zhang et al. (2016)
	Tapis oil	Oleic acid modified	10.7	Asadpour et al.

				(2016)
	Arabian crude oil	Oleic acid modified	11.9	Asadpour et al. (2016)
Pomelo Peel	Diesel		6.39	Chai et al. (2015)
	Diesel	Acetylation	18.91	Chai et al. (2015)
	Diesel	Styrene Modified	16.5	Chai et al. (2015)
	Lubrication oil		9.04	Chai et al. (2015)
	Lubrication oil	Acetylation	26.36	Chai et al. (2015)
	Lubrication oil	Styrene Modified	19.39	Chai et al. (2015)
	Diesel	Solvothermal treatment	27.98	Zou et al. (2016)
	Castor oil	Aerogel with hydrothermal carbonisation	5	Zhu et al. (2017)
	Sunflower oil	Aerogel with hydrothermal carbonisation	36	Zhu et al. (2017)
Coconut coir	Crude oil		4.4	Nduka et al. (2008)
	Diesel oil		3.64	Nduka et al. (2008)
	Kerosene		3.65	Nduka et al. (2008)

	Petrol oil		2.4	Nduka et al. (2008)
	Crude oil	carbonised	4.6	Nduka et al. (2008)
	Diesel oil	Carbonised	4.26	Nduka et al. (2008)
	Kerosene	Carbonised	3.2	Nduka et al. (2008)
	Petrol oil	Carbonised	2.52	Nduka et al. (2008)
	Crude oil		9.14	Yusof et al. (2015)
	Crude oil	Oleate Modified	12.38	Yusof et al. (2015)
	Crude oil	Octanoate Modified	9.56	Yusof et al. (2015)
	Crude oil	Acetylation - 1 % NBS	15.75	Teli and Vali (2013a)
	Crude oil	Butyl acrylate grafting	13.58	Teli et al. (2016)
	Vegetable oil		7.23	Ifelebuegu and Momoh (2015)
	Diesel oil		6.53	Ifelebuegu and Momoh (2015)
	Vegetable oil	Mixed with 50% human hair	7.92	Ifelebuegu and Momoh (2015)
	Diesel oil	Mixed with 50% human hair	6.6	Ifelebuegu and Momoh (2015)

	Cotton seed oil	Coco peat powder with Fe_3O_4	5.91	Yang et al. (2017)
	Paraffin oil	Coco peat powder with Fe_3O_4	5.14	Yang et al. (2017)
	Machine oil	Coco peat powder with Fe_3O_4	5.49	Yang et al. (2017)
	Silicon oil	Coco peat powder with Fe_3O_4	6.18	Yang et al. (2017)
Oil Palm Waste (Elaeis-Guineensis)	Tapis crude oil	Acetylation	6.8	Asadpour et al. (2016)
	Arabian crude oil	Acetylation	7	Asadpour et al. (2016)
	Engine oil		2.8	Idris et al. (2014)
	Crude oil	Lauric acid	1.18	Sidik et al. (2012)
Waste Paper	Crude oil	Aerogel functionalised with methyltrimethoxysilane	24.4	Nguyen et al. (2013)
	Various oils	Carbon microbelt aerogel	56 - 188	Bi et al. (2014)
	Various oils	Carbon aerogel	29 - 51	Han et al. (2015)
	Motor oil	Aerogel with methyltrimethoxysilane	95.5	Feng et al. (2015)
	Various oils	Aerogel with trimethoxylchlorosilane	12 - 22	Jin et al. (2015)
Bamboo Fibre	Waste motor oil	Aerogel	20.6	Wan et al. (2015)

	Pump oil	Aerogel	129	Yang et al. (2015)
	Machine oil	Aerogel	38	Jiao et al. (2016)
Bacteria Cellulose	Soya bean oil	Nanofibre aerogel	165	Wu et al. (2013)
	Pump oil	Nanofibre aerogel	140	Wu et al. (2013)
	Gasoline	Nanofibre aerogel	180	Wu et al. (2013)
	Diesel	Nanofibre aerogel	170	Wu et al. (2013)
	Various oils	Aerogel with trimethylchlorosilane treatment	86 - 185	Sai et al. (2015)
	Various oils	Nanofibre aerogel	64 - 120	Stefelova et al. (2017)
Azolla folliculoid Plant	Engine oil		11.29	Amin et al. (2015)
	Crude oil		5.22	Amin et al. (2015)
Biotemplanted Luffa Cylindrica	Engine oil	coated with Chitosan	2.38	Koseoglu (2016)
Raw Luffa	Heavy crude oil		9	Abdel Wahab (2014)
	Diesel oil		12	Abdelwahab (2014)
Pineapple Leaves	Crude oil		0.038	Cheu et al. (2016)
	Crude oil	Acetylation	0.091	Cheu et al. (2016)

Dacryodes Edulis Leaf	Crude oil		3.4	Naji et al. (2016)
	Crude oil	Acetylation	5	Naji et al. (2016)
Platanus(Plane)	Machine oil	Acetic Anhydride with NBS catalyst	46.41	Yang et al. (2016)
Winter Melon aerogel	Crude oil	Pyrolysed	25	Li et al. (2014)
Bleached Softwood fibre	Crude oil		6.5	Payne et al. (2012)
Eichhornia Cassipes	Crude oil	Treatment with polyamide epichlorohydrin resin	63	Yin et al. (2016)
	Diesel	Treatment with polyamide epichlorohydrin resin	77	Yin et al. (2016)
	Soybean oil	Treatment with polyamide epichlorohydrin resin	81	Yin et al. (2016)
	Lub oil	Treatment with polyamide epichlorohydrin resin	88	Yin et al. (2016)
	Silicon oil	Treatment with polyamide epichlorohydrin resin	94	Yin et al. (2016)

Table 2 Keratinous based biopolymers and their oil sorption capacities

Sorbent	Sorbate	Treatment /Modification	Sorption Capacity g/g	References
Human Hair	Crude oil		9.3	Ifelebuegu et al. (2015)
	Vegetable oil		8.1	Ifelebuegu et al. (2015)
	Diesel oil		7.92	Ifelebuegu et al. (2015)
Chicken Feathers	Vegetable oil		7.7	Ifelebuegu and Chinonyere (2015)
	Crude oil		6.1	Ifelebuegu and Chinonyere (2015)
	Diesel oil		4.1	Ifelebuegu and Chinonyere (2015)
Wood Fibre	Machine oil		33-43	Ognjanovic and Rajakovic (2008)
	Machine oil	Recycled non-woven	15-19	Ognjanovic and Rajakovic (2008)
	Base oil		14	Radetic (2003)
	Diesel oil		13	Radetic (2003)
	Crude oil		12	Radetic (2003)
	Diesel oil	Recycled non-woven	9.62	Radetic et al. (2008)
	Crude oil	Recycled non-woven	11.06	Radetic et al. (2008)
	Base oil		12.98	Radetic et al. (2008)
	Vegetable oil		13.16	Radetic et al. (2008)

	Motor oil		15.8	Radetic et al. (2008)
Silk Fibre	Crude oil		68.7 - 85.5	Annunciado et al. (2005)
	Motor oil		42 - 52	Moriwaki et al. (2009)
	Vegetable oil		37 - 60	Moriwaki et al. (2009)

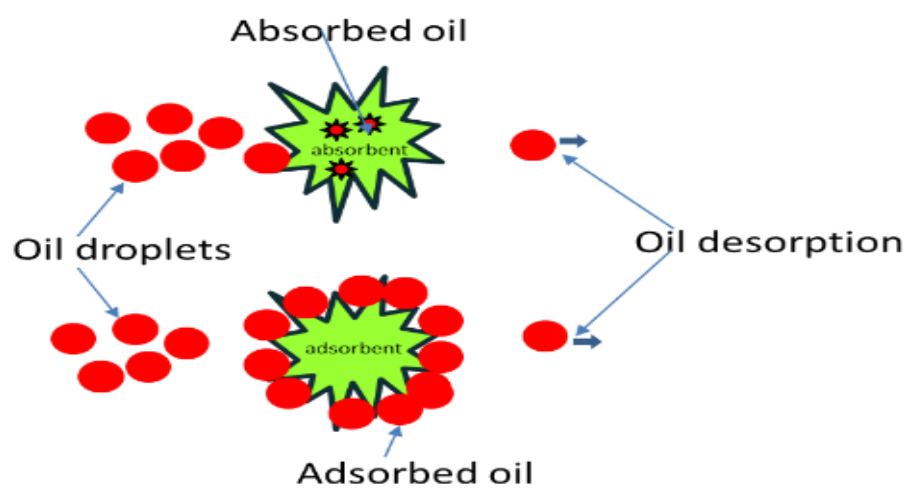


Figure 1. Oil absorption and adsorption processes

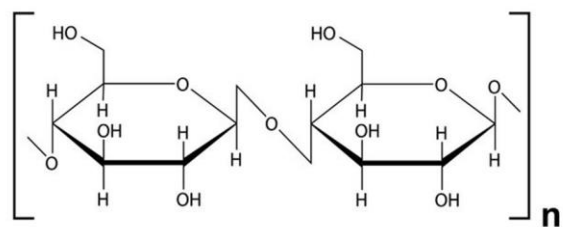


Figure 2. Chemical structure of cellulose

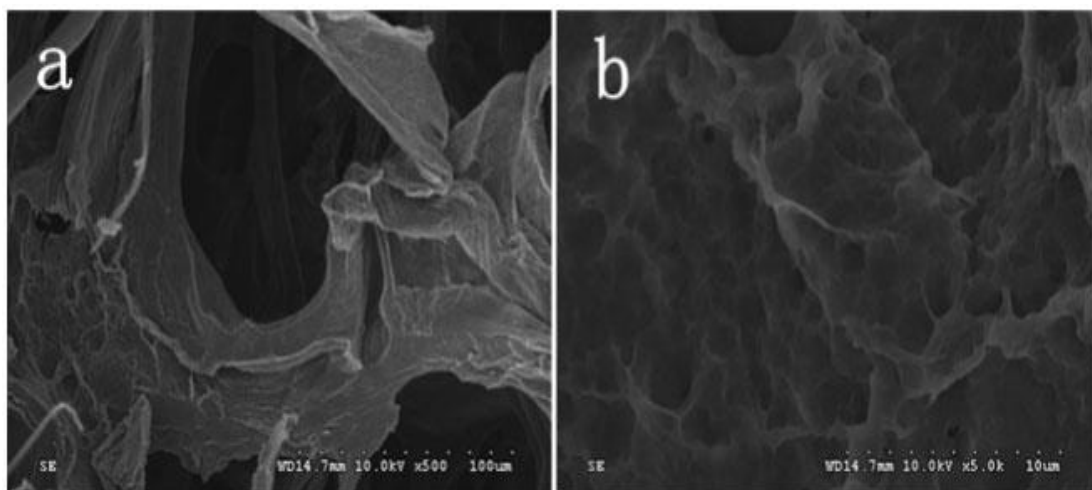


Figure 3. SEM image of cellulose aerogel (a) inner pores (b) surface pores (Shi et al., 2015) © Taylor and Francis.